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Production of long-lived exotic radionuclides for nuclear physics experiments

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Outline

- Objective of the project
- ERAWAST plan
- Isotope production sources
  - Copper beam dump
  - Lead targets from SINQ
- Summary and Outlook

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Objective of the project ERAWAST

(Exotic Radionuclides from Accelerator Waste for Science and Technology)

Application of exotic long-lived isotopes from accelerator waste for scientific purposes

Examples:

^{7}Be (50.8d), ^{10}Be (1.6·10^{6}y), ^{26}Al (7.2·10^{5}y), ^{44}Ti (60.4y), ^{53}Mn (3.7·10^{6}y), ^{60}Fe (1.5 ·10^{6}y), ^{146}Sm (1.03 ·10^{8}y), ^{182}Hf (9 ·10^{6}y) , ^{129}I (1.56 ·10^{7}y)

Development of a long-term international collaboration between

- Nuclide production facilities
- Basic physics research
- Nuclear Data for P&T
- Nuclear astrophysics
- AMS measurement groups
- Pharmaceutical chemistry
- Nanotechnology

ERAWAST – plan

1. **Existing accelerator waste material**
   Copper beam dump irradiated at the 590-MeV proton beam station at PSI, dismounted about 15 years ago
   ^{26}Al, ^{59}Ni, ^{53}Mn, ^{60}Fe, ^{44}Ti or others can be separated
   other irradiated materials like carbon (^{10}Be), stainless steel or concrete are also available
   1 PostDoc position financed from PSI for the development of separation in a technological scale now available
2. **Target material from the SINQ facility**
   Two irradiated lead targets from the spallation source are available. Heavier isotopes like ^{182}Hf, ^{129}I or several rare earth elements (e.g. ^{146}Sm, several Dy isotopes) can be obtained. In principle, targets from the SINQ will be available every second year.
3. **Special irradiations**
   The SINQ facility offers the possibility to irradiate materials with 590 MeV protons at special positions. Tended experiments for isotope production can be offered.

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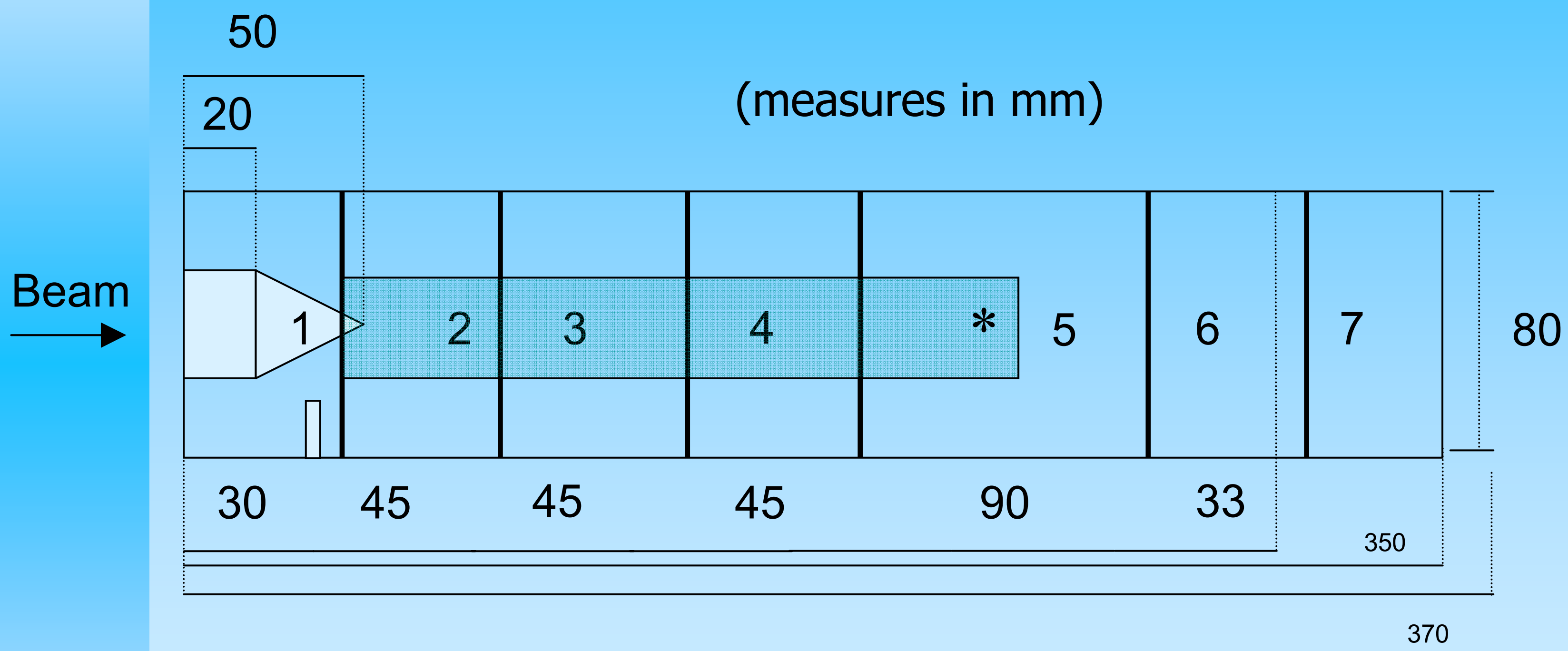
Characteristics of the copper beam dump

- Beam stop from the former BMA station
- Operated from 1980-1992, dismounted in 1993
- 0.1 Ah total beam dose (590 MeV protons)
- copper cylinder of ~ 10 kg; diameter 80mm
- Sample taking from several parts by drilling

Schematic view of the beam dump



* - Area of drilling Ø 20mm

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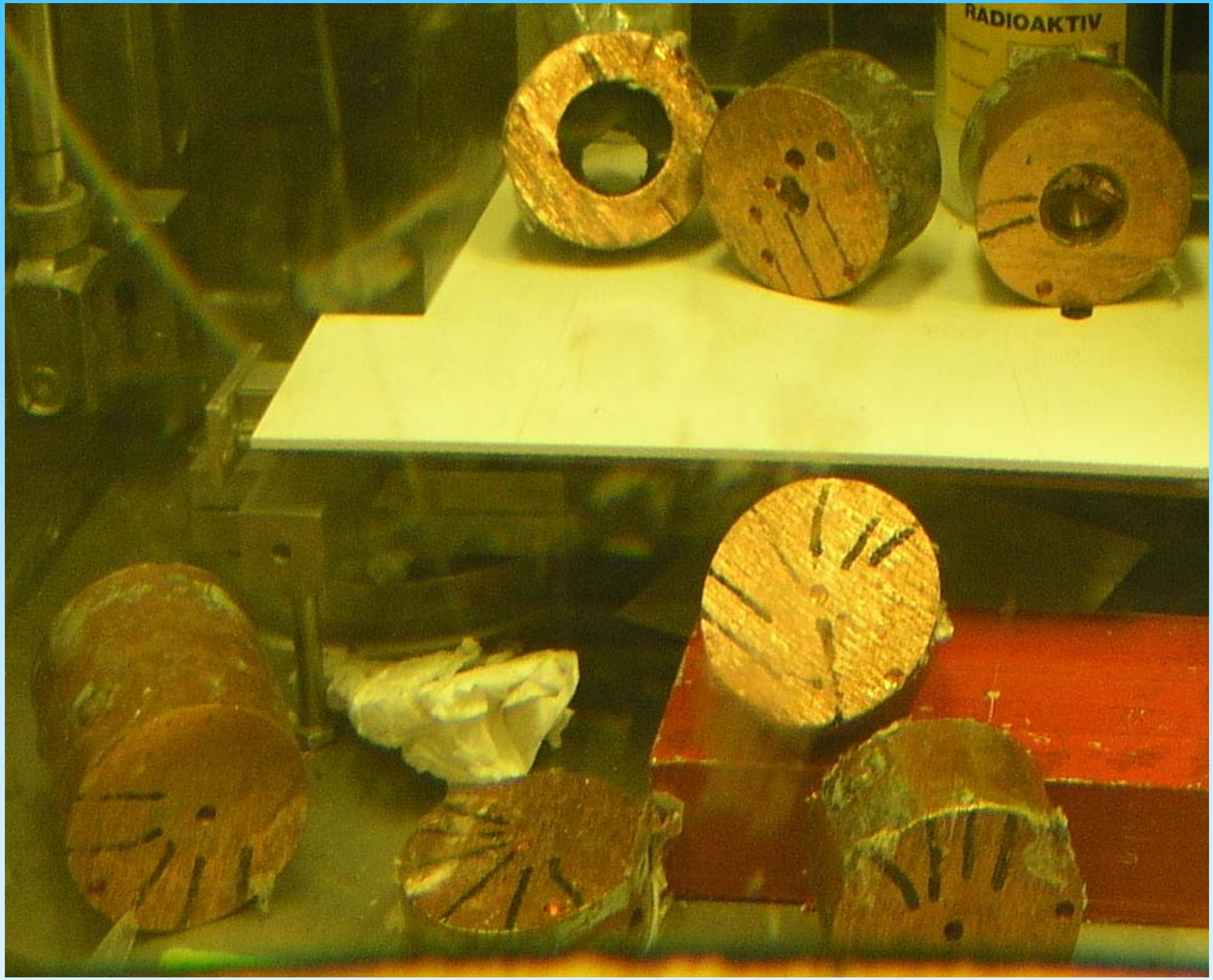
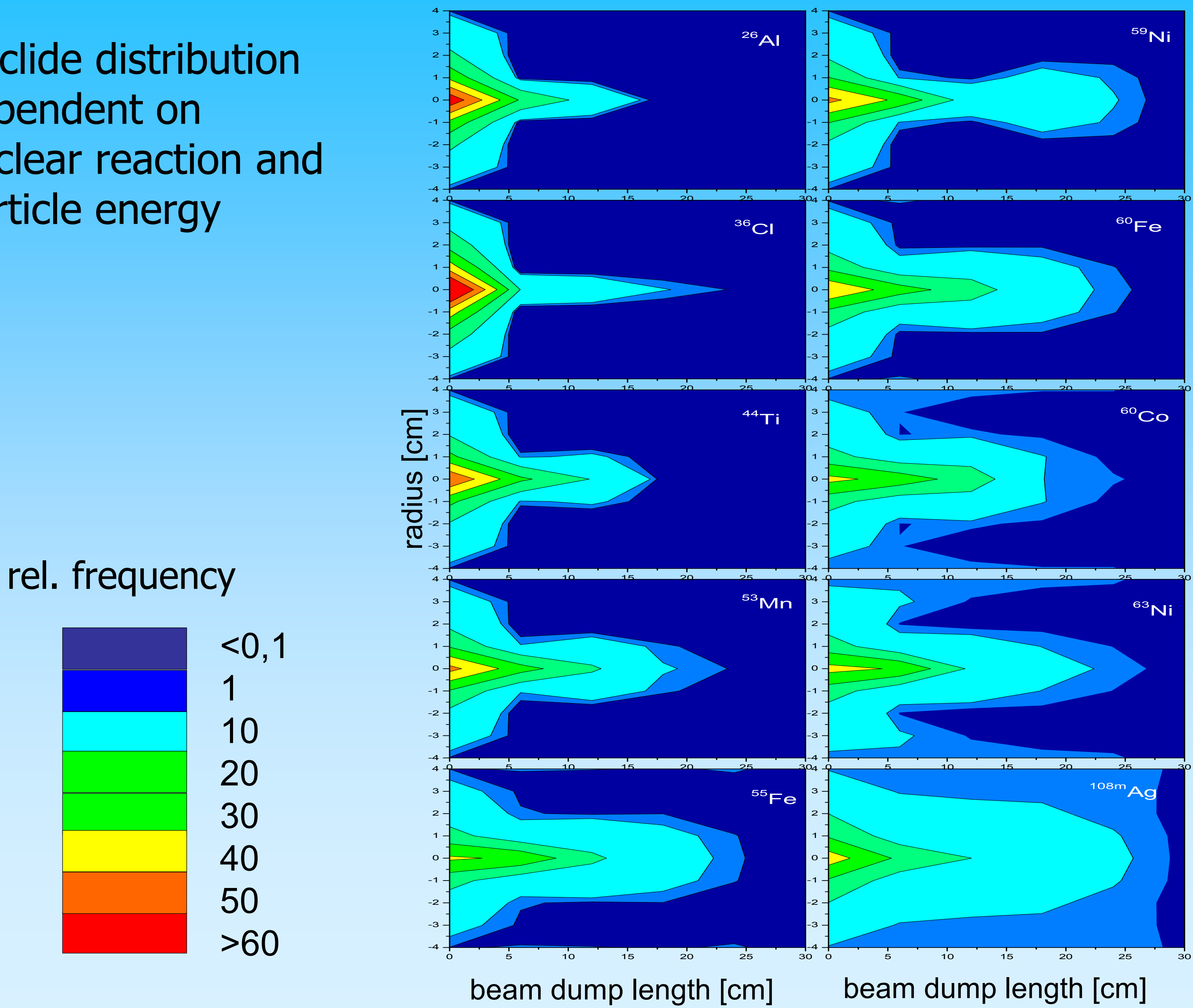
Before drilling



Sample	^{36}Cl [Bq/g]	^{60}Fe [Bq/g]	^{26}Al [Bq/g]	^{108m}Ag [Bq/g]	^{53}Mn [Bq/g]	^{44}Ti [kBq/g]	^{63}Ni [kBq/g]	^{55}Fe [kBq/g]	^{59}Ni [kBq/g]	^{60}Co [kBq/g]
C6.2.1	**1281**	**84.7**	**140**	**62.58**	**6900**	**1616.0**	**34151.1**	**42450.3**	**127.0**	**49957.1**
C6.2.2	0.30	-	0.01	1.35	0.6	0.2	217.7	108.6	0.13	**111.6**
C6.3.1	**279**	**58.5**	**56**	**37.27**	**4310**	**740.8**	**36566.7**	**44136.3**	**125.0**	**37969.6**
C6.3.2	0.19	3.3	1	9.19	112	18.6	2006.0	2562.0	6.62	**2691.8**
C6.3.3	0.02	1.9	0.2	9.22	117	1.5	1109.7	1552.4	2.62	**1239.4**
C6.3.4	0.01	0.5	0.03	1.69	6	0.6	1841.6	257.0	0.76	**663.2**
C6.3.5	0.35	0.5	0.01	1.06	4.9	0.4	706.2	154.7	0.47	**438.8**
C6.4.1	**173**	**52.2**	**41**	**27.70**	**3600**	**778.1**	**16776.3**	**26590.4**	**-**	**47256.0**
C6.4.2	0.24	-	0.008	1.27	2.1	-	799.1	132.5	-	**505.5**
C6.5.1	**79**	**20.2**	**3**	**13.64**	**998**	**95.0**	5764.1	**11520.4**	**25.7**	**10091.9**
C6.5.2	0.27	-	0.01	1.80	2.0	-	545.6	157.8	0.42	**415.0**
C6.6.1	0.13	**0.7**	0.012	**3.93**	**10.4**	-	1005.7	**287.6**	**6.94**	**459.0**
C6.6.2	0.08	-	0.0019	0.94	0.6	-	233.2	127.8	-	**169.8**
C6.7.1	0.08	**0.7**	0.005	**1.34**	1.4	-	170.2	**350.7**	-	**148.6**
C6.7.2	0.04	-	0.0013	0.86	0.5	-	118.8	233.6	-	**91.1**

Nuclide distribution dependent on nuclear reaction and particle energy



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Estimation of available radionuclides

(no separation)

Basis: central drilling with 2 cm diameter
500g copper
Average of all 4 central values
Average of the central value and from 1 cm distance
„Safety factor" of 2

^{44}Ti: 100 MBq (10^{18} atoms)
^{53}Mn: 500 kBq (10^{19} atoms)
^{26}Al: 7 kBq (10^{17} atoms)
^{60}Fe: 5 kBq (10^{17} atoms)
^{59}Ni: 8 MBq (10^{19} atoms)
(^{60}Co: 5 GBq)

All these radionuclides can be provided without carrier, but some of them contain other long-lived isotopes (^{55}Fe/^{63}Ni)

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Preparation of ^{60}Fe-samples

- Chemical separation of the iron fraction from samples of the copper beam-dump (590 MeV protons, beam dosis ca. 0.1Ah); ~10^{15}-10^{16} atoms
- Basic research:
  - Determination of the half-life of ^{60}Fe (collaboration with TU Munich, Germany)
  - Cross section measurement of the reaction $^{60}Fe(n,\gamma)^{61}Fe$ (collaboration with FZK, Germany)
- Applied research: Preparation of standard solutions for accelerator mass spectrometry, biomedical investigations, astrophysics

Chemical separation of ^{60}Fe

Special Problem:

^{60}Fe (1.5·10^6 y) $\xrightarrow{\beta^-}$ ^{60m}Co (10.5 min) $\xrightarrow{\gamma}$ ^{60}Co (5.3 y) $\xrightarrow{\gamma,\beta^-}$ ^{60}Ni (stable)

^{60}Fe: no γ radiation, low β–energy

Measurement of the increase of the Co-daughter
Measurement of the ^{61}Fe production (1027/1205 keV)
→ very good chemical separation from Co necessary

- Dissolution of Cu chips(beam dump) in 7 M HNO_3
- Evaporation to dryness
- Dissolution in 7 M HCl
- + 5 mg Fe^{3+} and 5 mg Co^{2+} as carrier
- Extraction with diisopropylether
- Aqueous phase: Ni, Co, Cu, organic phase: Fe
- Back extraction with 0.1 M HCl, repetition of procedure
- Additional purification by precipitation of $Fe(OH)_3$
- Result: $3.8 \cdot 10^{15}$ (TUM) / $1.2 \cdot 10^{16}$ (FZK) ^{60}Fe atoms, decontamination factor (Co) > 10^8 (0.3 Bq)

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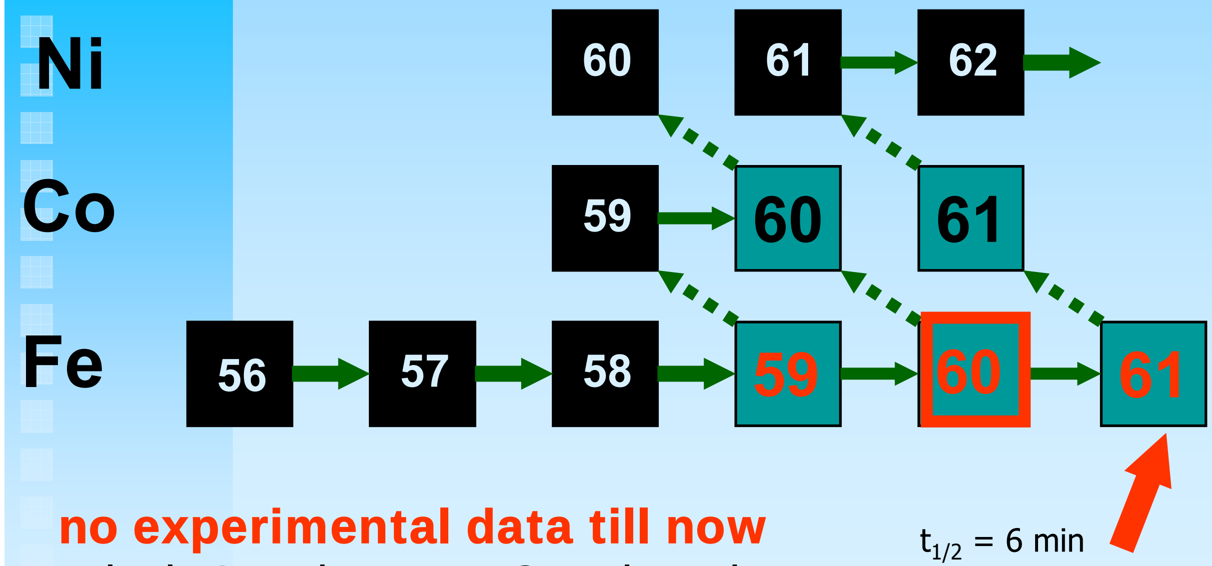
A key reaction for understanding the composition of the Early Solar System (ESS): ^{60}Fe(n, g)^{61}Fe

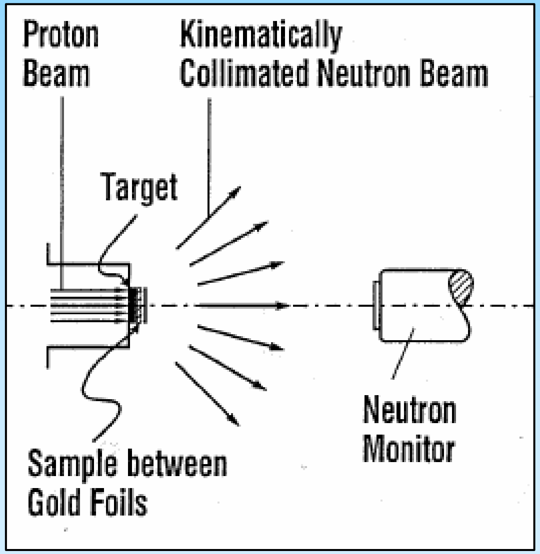
neutron flux during C-shell burning in massive stars sufficient to bridge ^{59}Fe ($t_{1/2}$ = 44 d)
stellar production of **^{60}Fe** determined by (n, g) rates for production and destruction
Cross section for destruction of **^{60}Fe** measured for the first time!

- ~10^{16} atoms extracted from a copper beam dump at PSI for target preparation
- detection of product nucleus by Karlsruhe activation technique



no experimental data till now
calculations between 3 and 7 mbarn
are uncertain by a factor of 3!

$t_{1/2}$ = 6 min
E_g = 298, 1027, 1205 keV
I_g = 22, 43, 44(5)%



Activation technique: neutron production via ^{7}Li(p,n)^{7}Be reaction at *Ep* = 1912 keV

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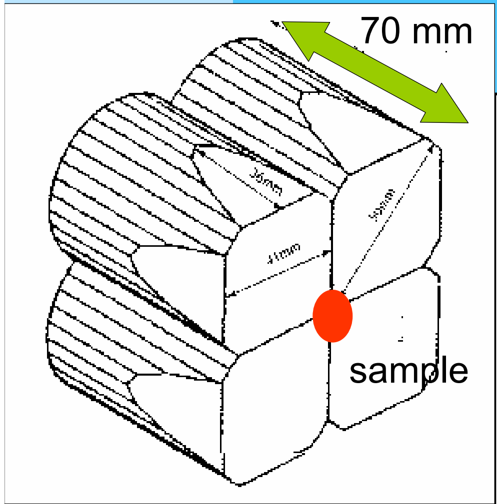
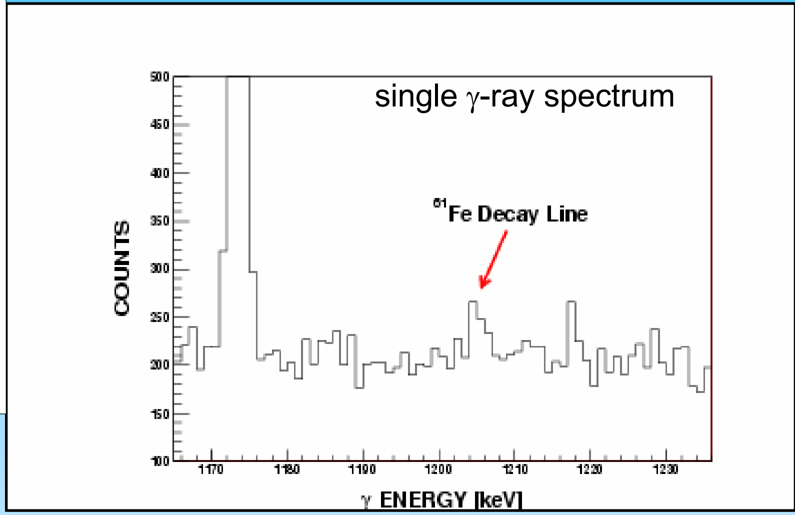
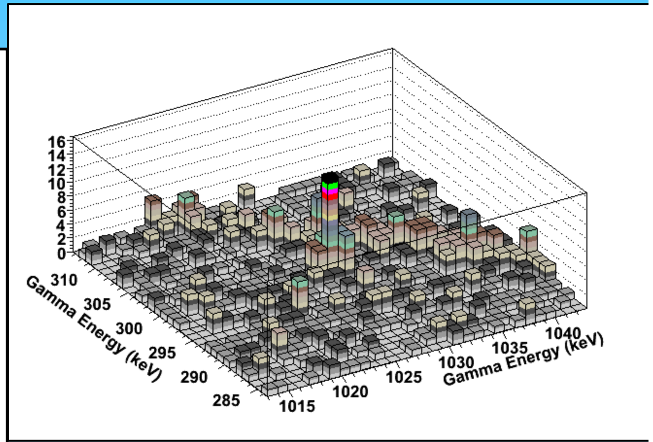
Preliminary result

^{60}Fe sample irradiated 64 times for 15 min, induced activity counted for 10 min



γ-counting: 2 Ge Clovers face to face





γ-ray spectrum of coincident events

- total number of ^{60}Fe atoms = $7.8 \cdot 10^{15}$ or 777 ng with $T_{1/2}$ (^{60}Fe) = $1.5 \cdot 10^{6}$ years
- total number of capture events = 118 (single) and 17 (coinc)
- time-integrated neutron flux = $1.7 \cdot 10^{14}$

$^{60}Fe(n, \gamma)^{61}Fe$ cross section @ kT = 25 keV

$\langle\sigma\rangle = 10.9 \pm 3.1_{syst} \pm 1.5_{stat}$ mbarn

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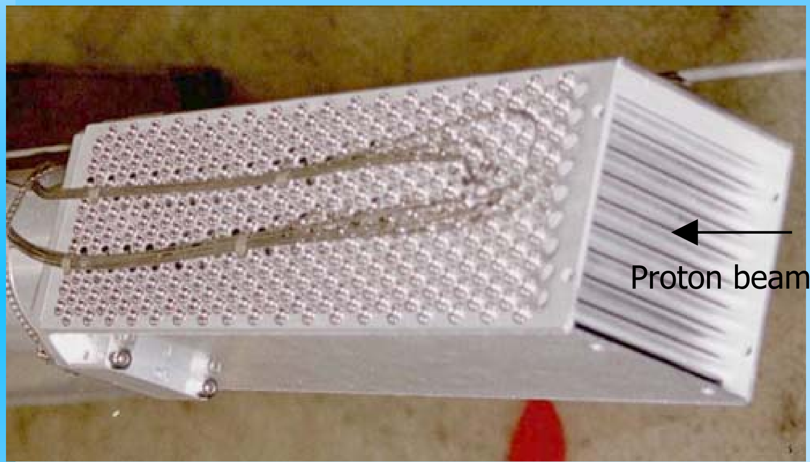
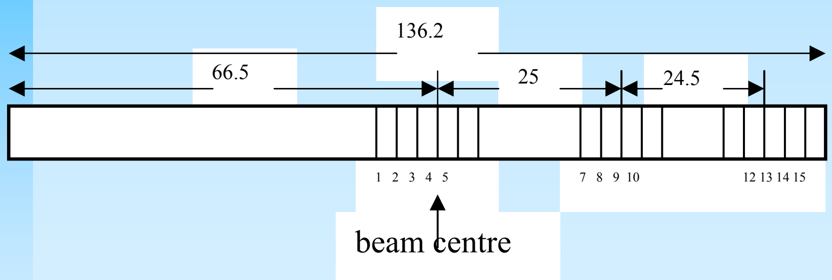
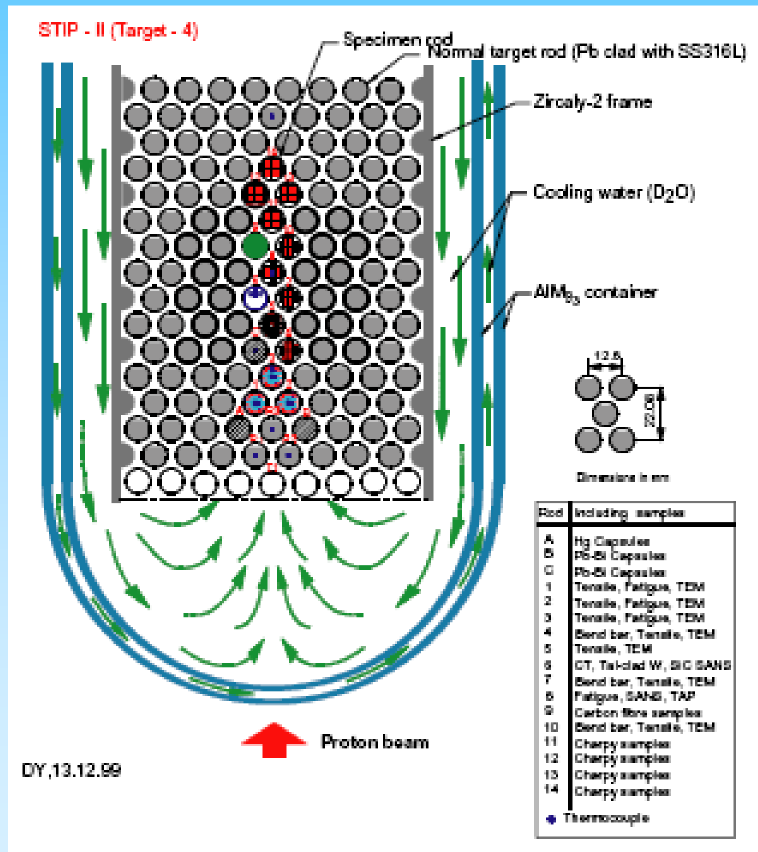
Lead targets from SINQ

2 Samples from target 4, 2 years operation; EOB 1999







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Analysis results

	D9 [Bq/g]	D14 [Bq/g]		D9 [Bq/g]	D14 [Bq/g]
^{207}Bi	$3.00 \cdot 10^{7}$	$1.01 \cdot 10^{7}$	^{106}Ru	$4.83 \cdot 10^{6}$	$3.91 \cdot 10^{6}$
$^{172}Lu/^{172}Hf$	$2.00 \cdot 10^{7}$	$5.41 \cdot 10^{7}$	^{110m}Ag	$1.29 \cdot 10^{6}$	$3.93 \cdot 10^{5}$
^{173}Lu	$2.76 \cdot 10^{7}$	$4.30 \cdot 10^{7}$	^{125}Sb	$1.32 \cdot 10^{6}$	-
$^{194}Au/^{194}Hg$	$1.86 \cdot 10^{7}$	$3.13 \cdot 10^{6}$	^{133}Ba	$2.8 \cdot 10^{6}$	$7.94 \cdot 10^{5}$
^{102}Rh	$5.53 \cdot 10^{6}$	$1.44 \cdot 10^{5}$	$^{44}Sc/^{44}Ti$	$8.00 \cdot 10^{4}$	$2.84 \cdot 10^{4}$
$^{202}Tl/^{202}Pb$	$4.80 \cdot 10^{5}$	$1.87 \cdot 10^{5}$	^{108m}Ag	$3.75 \cdot 10^{5}$	$1.56 \cdot 10^{4}$
^{60}Co	$3.67 \cdot 10^{6}$	$1.40 \cdot 10^{6}$	$^{194}Os/^{194m}Ir$	$2.61 \cdot 10^{4}$	-
^{54}Mn	$2.29 \cdot 10^{5}$	$7.01 \cdot 10^{4}$	^{26}Al	0.5	0.2
^{58}Co	$1.55 \cdot 10^{6}$	$9.47 \cdot 10^{5}$	^{36}Cl	$9.5 \cdot 10^{1}$	$4.8 \cdot 10^{1}$
^{55}Fe	$8.73 \cdot 10^{7}$	$5.99 \cdot 10^{7}$	^{63}Ni	$6.30 \cdot 10^{8}$	$4.52 \cdot 10^{8}$

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Summary

- ERAWAST-community established (appr. 30 partners, growing number), new proposal for Network Programme
- First samples provided: ^{10}Be (GSI), ^{60}Fe (TUM; FZK), ^{26}Al, ^{44}Ti (Uni Mainz; Edinburgh)
- Up to 10^{18} atoms of ^{10}Be available (separated)
- 10^{16-19} atoms of several radionuclides (^{26}Al, ^{60}Fe, ^{59}Ni, ^{53}Mn, ^{44}Ti) available (not yet separated)
- First experiment with ^{60}Fe successful: neutron capture cross section; half-life measurement underway
- PostDoc for subsequent separation from Cu in a Hot-cell available
- Analytical work on Pb-targets ongoing, heavier isotopes can in principle be provided
- More Cu-, Pb- and C-samples available
- Possibilities for other irradiation positions (SINQ, beam dumps, collimators)
- Problem of mass separation still unsolved